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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of : Confirmation No. 7513
Nobutaka AKASHI et al. : Attorney Docket No. 10893.0064USWO
Serial No. 10/580,052 : Group Art Unit 1794
Filed May 19, 2006 : Examiner Dawn Garrett

ORGANO-ELECTRONIC FUNCTIONAL MATERIAL AND USE THEREOF

DECLARATION UNDER 37 CFR 1.132

I, Nobutaka AKASHI, declare as follows:

I was graduated from Graduate School of Science, Himeji Institute of Technology (currently University of Hyogo), Hyogo, in March, 1996, and I have been employed by BANDO CHEMICAL INDUSTRIES, LTD. since April, 1996. I am at present a researcher at Department of Products Development, R & D Center, Kobe-shi. I have been engaged in research of organic electroluminescence as from 2001 up to now.

1. Object of the Experiments

I have conducted the following experiments to compare the performance of an organic electroluminescence element according to the present invention in which a hole transporting layer is formed using 1,3,5-tris(N-(4'-methyl-4-biphenyl)-N-(p-tolyl)amino)benzene (p-MBTAB) with that of an organic electroluminescence element in which a hole transporting layer is formed using N,N,N',N',N'',N''-hexakis-(4-methylphenyl)-1,3,5-triaminobenzene (p-MPTAB) mentioned in JP 11-176572A..

2. Experiments

(1) Synthesis of p-MBTAB

2.0 g of 1,3,5-tris(p-tolylamino)benzene, 7.0 g of 4-iodo-

4'-methylbiphenyl, 6.9 g of potassium carbonate, 1.0 g of copper powder, 0.7 g of 18-crown-6 (i.e., 1,4,7,10,13,16-hexaoxacyclooctadecane) and 15 mL of mesitylene (reaction solvent) were placed in a 100 mL capacity glass flask and the reaction was carried out at a temperature of 170°C for 15 hours under a nitrogen atmosphere.

After the reaction, the resultant reaction mixture was extracted with toluene and the toluene solution was subjected to silica gel chromatography to fractionate the reaction product. The reaction product was then purified by recrystallization and then by sublimation to provide the desired p-MBTAB. The yield was 15.6%.

p-MBTAB was found to have a molecular weight of 892 as measured by mass analysis, and a glass transition temperature (T_g) of 103.4°C as measured by differential scanning calorimetry.

(2) Preparation of organic electroluminescence element according to the invention

A sheet of plate glass having an ITO coating on one face (available from Sanyo Vacuum K.K.) was subjected to ultrasonic cleaning using acetone and then steam cleaning using methanol, followed by irradiation with ultraviolet rays by using a low-pressure mercury lamp for 10 minutes. Immediately after the irradiation, 4,4',4''-tris(N-(2-naphthyl)-N-phenyl-amino)triphenylamine (2-TNATA) was vapor deposited to form a hole injecting layer 60 nm thick and then p-MBTAB was vapor deposited to form a hole transporting layer 40 nm thick in this order on the ITO coating by using a vacuum deposition apparatus. Subsequently, an emission layer 75 nm thick was formed of tris(8-quinolinol)aluminum (Alq₃) on the hole transporting layer, and then a lithium fluoride layer 0.75 nm thick and an aluminum layer 100 nm thick were layered in this order on the emission layer to form a cathode, thereby providing an organic electroluminescence element.

(3) Synthesis of p-MPTAB

10.0 g of 4,4'-ditolylamine, 4.8 g of 1,3,5-tribromobenzene, 8.9 g of

sodium t-butoxide and 150ml of xylene (reaction solvent) were placed in a 200 mL capacity three-necked glass flask, and the resulting mixture was heated to a temperature of 120°C with stirring under a nitrogen atmosphere. Then, 0.034 g of palladium acetate and 0.062 g of tri-t-butylphosphine were added to the mixture, and the reaction was carried out at a temperature of 120°C for five hours with stirring.

250ml of ethanol was added to the reaction mixture to cease the reaction, and the resulting solid was collected by filtration. The solid was purified by silica gel chromatography, and then by recrystallization from a mixed solvent composed of 110 ml of toluene and 30 ml of ethanol to provide 8.3 g of the desired p-MPTAB. The yield was 81%.

p-MPTAB was found to have a molecular weight of 664 as measured by mass analysis, and a glass transition temperature (T_g) of 67°C as measured by differential scanning calorimetry.

(4) Preparation of organic electroluminescence element as a comparative example

A sheet of plate glass having an ITO coating on one face (available from Sanyo Vacuum K.K.) was subjected to ultrasonic cleaning using acetone and then steam cleaning using methanol, followed by irradiation with ultraviolet rays by using a low-pressure mercury lamp for 10 minutes. Immediately after the irradiation, 4,4',4''-tris(N-(2-naphthyl)-N-phenylamino)triphenylamine (2-TNATA) was vapor deposited to form a hole injecting layer 60 nm thick and then p-MPTAB was vapor deposited to form a hole transporting layer 40 nm thick in this order on the ITO coating by using a vacuum deposition apparatus. Subsequently, an emission layer 75 nm thick was formed of tris(8-quinolinol)aluminum (Alq₃) on the hole transporting layer, and then a lithium fluoride layer 0.75 nm thick and an aluminum layer 100 nm thick were layered in this order on the emission layer to form a cathode, thereby providing an organic electroluminescence element.

(5) Performance of organic electroluminescence elements of the invention and as a comparative example

Each of the organic electroluminescence elements of the invention and as a comparative example was stood at a temperature of 90°C in an oven for one hour, allowed to cool to room temperature (25°C), and then power efficiency of emission, current efficiency of luminance, emission luminance, and voltage applied across the cathode and the anode when the organic electroluminescence element was driven at a fixed current density of 25 mA/cm².

After the measurement, the organic electroluminescence element was again stood at a temperature of 90°C in an oven for one hour, allowed to cool to room temperature, and then the same measurement as mentioned above was made. In this way, the organic electroluminescence element was heated and then subjected to the measurement six times. The results are shown in Fig. 1 to Fig. 4 in which the change of performance of organic electroluminescence element when it was driven at a fixed current density of 25 mA/cm² before and after it was heated for a predetermined period of time at a temperature of 90°C.

Fig. 1 shows power efficiency of emission vs. heating time; Fig. 2 shows current efficiency of luminance vs. heating time, Fig. 3 shows emission luminance vs. heating time, and Fig. 4 shows voltage applied across the electrodes to drive the element at a fixed current density of 25 mA/cm² vs. heating time.

(6) Results

The organic electroluminescence element of the invention was found to have substantially the same power efficiency of emission, current efficiency of luminance, and emission luminance after heating at 90°C for six hours as those the organic electroluminescence element had before heating. However, the organic electroluminescence element as a comparative example was found to be remarkably reduced in power efficiency of emission, current efficiency of luminance, and emission luminance just after the first one hour heating.

The organic electroluminescence element of the invention was able to

be driven at a fixed current density of 25 mA/cm² by applying a voltage of 8.26 V across the electrodes, and was found to be able to be driven by applying almost the same voltage even after it was heated at a temperature of 90°C for six hours.

The organic electroluminescence element as a comparative example was able to be driven at a fixed current density of 25 mA/cm² by applying almost the same voltage before it was heated as the organic electroluminescence element of the invention. However, after first one hour heating, the organic electroluminescence element as a comparative example was found to need a voltage as large as 8.92 V so that it was driven at a fixed current density of 25 mA/cm². After it was heated for six hours, the organic electroluminescence element as a comparative example was found to need a voltage as large as 9.15 V so that it was driven at a fixed current density of 25 mA/cm².

From the above results I conclude that the organic electroluminescence element of the invention has much more improved heat-resistance than that of the comparative example because the former has a hole transporting layer formed with p-MBTAB of the invention.

I further declare that all statements made herein of his own knowledge are true, and that all statements on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Feb. 24, 2009
Date

Robutaka Akashi
Signature

FIG. 1

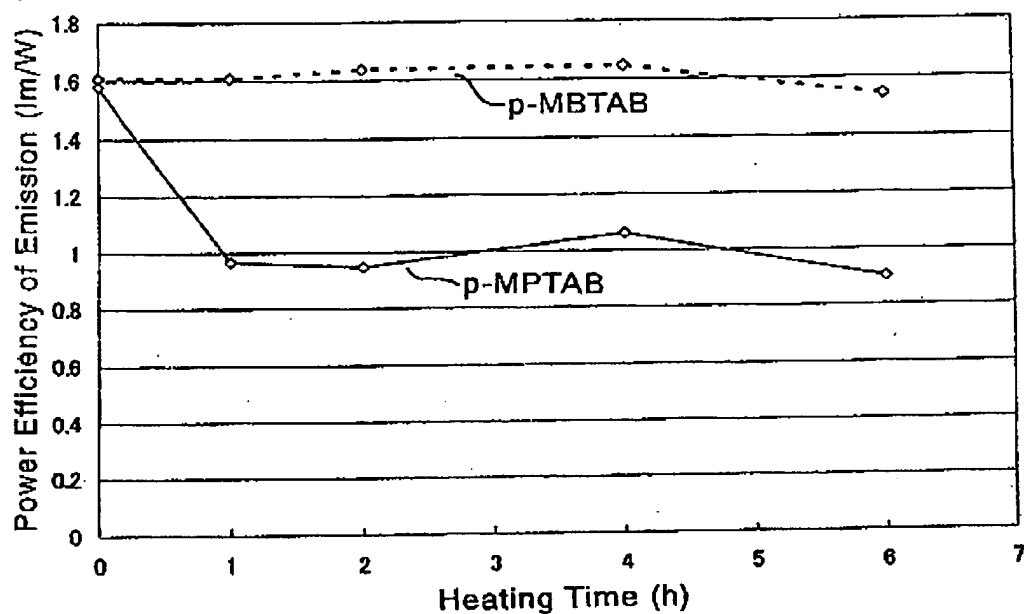


FIG. 2

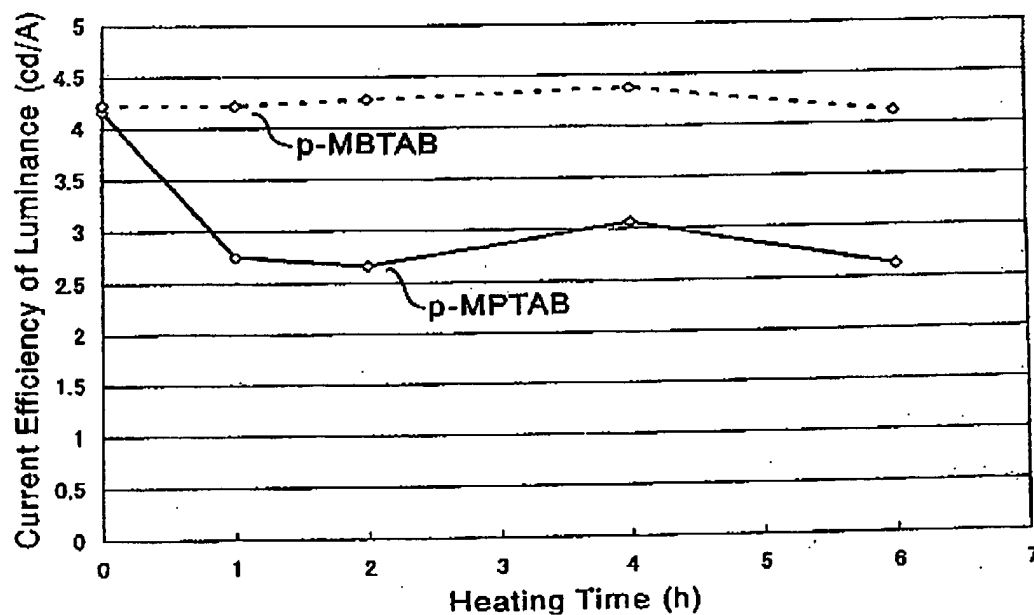


FIG. 3

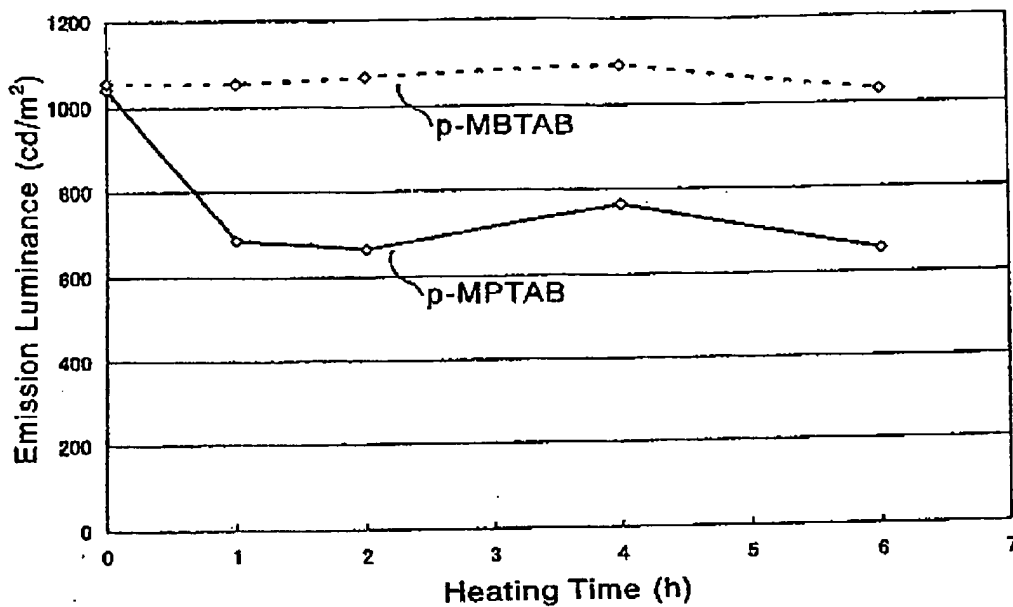


FIG. 4

